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E. J. Reed, M. Soljacic, R. Gee, J. D. Joannopoulos

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PREDICTION OF COHERENT OPTICAL RADIATION FROM SHOCK WAVES IN POLARIZABLE CRYSTALS

Evan J. Reed[*†], Marin Soljačić[†], Richard Gee[*] and J. D. Joannopoulos[†]

[*]*Lawrence Livermore National Laboratory, Livermore, CA 94551*
[†]*Center for Materials Science and Engineering and Research Laboratory of Electronics, Massachusetts Institute of Technology, Cambridge, MA 02139*

Abstract. We predict that coherent electromagnetic radiation in the 1-100 THz frequency range can be generated in crystalline materials when subject to a shock wave or soliton-like propagating excitation. To our knowledge, this phenomenon represents a fundamentally new form of coherent optical radiation source that is distinct from lasers and free-electron lasers. General analytical theory and molecular dynamics simulations demonstrate coherence lengths on the order of mm (around 20 THz) and potentially greater. The emission frequencies are determined by the shock speed and the lattice constants of the crystal and can potentially be used to determine atomic-scale properties of the shocked material.

Keywords: shock wave, optical emission, molecular dynamics, sodium chloride,
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INTRODUCTION

The invention of lasers in 1958 made possible a staggeringly wide range of applications. The key characteristic of lasers that enables many of these applications is the fact that they are sources of *coherent* light. Almost 50 years later, very few distinct ways to generate coherent light have been realized. These include: "traditional" lasers based on stimulated emission and free-electron lasers, [1] each with its own unique practical advantages and disadvantages. This work presents what we believe is a new source of coherent optical radiation that is fundamentally distinct from lasers and free-electron lasers. We perform analytical theory and molecular dynamics simulations that predict that weak yet measurable *coherent* light can be observed emerging from a shocked polarizable crystal, typically in the range 1-100 THz. The *periodicity of the crystalline lattice* is the origin of the coherence of emitted radiation *rather than the "coherence" of the source generating the shock wave.*

This effect is predicted to be observable in a wide variety of material systems under realizable shock wave conditions. In this work, we consider shock waves in NaCl. Some experiments on shocked single NaCl crystals have been reported. [2, 3] To our knowledge, coherent emission has never been observed because a shocked crystal is not an obvious system to discover (and hence look for) coherent radiation and the radiation is in a portion of the electromagnetic spectrum that is usually not observed in such experiments.

ANALYTICAL THEORY

As a shock propagates through a polarizable crystal, a change in polarization can be induced which yields a time-dependent polarization current (even in materials with no static polarization). While it is not surprising that radiation should be emitted from the polarization currents induced by the shock, it is unexpected that this emission should be of a coherent nature. The coherent property of the radiation is the

subject of this work. The frequencies of the polarization current are associated with the temporal period of the shock propagating through a single lattice unit of the crystal. The periodicity of the crystal lattice is the true origin of the coherent emission.

In this section we use an analytical approach to demonstrate the coherent nature of the emitted radiation. To represent the material, a polarizable element $P_n(t)$ that exists on each lattice point n located at $x = na$ obeys the equation,

$$\frac{d^2P_n(t)}{dt^2} = \mu_n(t)E_n(t) - \Omega_n(t)^2P_n(t) - f_n(t) - \gamma(t)\frac{dP_n(t)}{dt}. \quad (1)$$

Here, $\mu_n(t)$ is a polarizability-related parameter, $\Omega_n(t)$ is the resonant frequency of the n^{th} polarizable element, and $\gamma(t)$ is an absorption term. The term $f_n(t)$ represents coupling to the shock wave and is a forcing term that generates shock-induced changes in polarization. Ω_n is the local transverse optical mode frequency (ω_T) ranging from $10^{13}s^{-1}$ for phonons in ionic crystals to $10^{15}s^{-1}$ and higher for electronic excitations. Equation 1 can model many polarizable materials when combined with Maxwell's equations. In the case where $\Omega_n(t)$ and $\mu_n(t)$ are time-independent and $f_n = 0$, Equation 1 produces the usual polariton dispersion relation. [4]

To determine emission characteristics of this shocked polarizable material, we perform a symmetry analysis of the classical equations of motion of the system. There exists a time and space translational invariance of this system that gives rise to a Bloch-like property for the fields. Define a space and time translation operator $\hat{T}_m$ such that $\hat{T}_m g_n(t) \equiv g_{n-m}\left(t - m\frac{a}{\mathrm{v_s}}\right)$. In the shock wave, suppose that the polarizable elements have the property that $\hat{T}_m\mu_n(t) = \mu_n(t)$, $\hat{T}_m\Omega_n(t) = \Omega_n(t)$, $\hat{T}_m f_n(t) = f_n(t)$, and $\hat{T}_m\gamma_n(t) = \gamma_n(t)$. Comparison of the fields in Equation 1 and Maxwell's equations with and without the application of $\hat{T}_m$ leads to the result that the electric field E must be of the form,

$$E = \sum_k e^{ik(x-\mathrm{v_s}t)} \sum_\ell E'_{k,\ell} e^{-2\pi i\ell\frac{\mathrm{v_s}}{a}t} \quad (2)$$

where ℓ is an integer and k is a wave vector and H has a similar form. The Bloch-like property of the fields yields a condition on the radiation emitted by the shock wave. Possible frequencies in the fields in Eq. 2 are,

$$\omega_1 = k_1\mathrm{v_s} + 2\pi\ell\frac{\mathrm{v_s}}{a} \quad (3)$$

where subscript 1 denotes the output radiation. Possible emission frequencies into the pre-shock and post-shock materials are those for which Eq. 3 and $\omega(k_1)$ for the pre-shock and post-shock materials have common solutions, respectively. The emission frequencies for $\ell \neq 0$ are highly anomalous since $\frac{2\pi}{a} >> k_1$. Since the lattice constant a is typically several orders of magnitude smaller than the wavelength of optical light, these frequencies for $\ell \neq 0$ are larger than a typical Doppler shift ($k_1\mathrm{v_s}$) by several orders of magnitude. The confinement of the emitted radiation to discrete frequencies demonstrates the coherent nature in this model.

COMPUTATIONAL EXPERIMENTS

In this section, we numerically explore the light generated by a shocked polarizable material by performing molecular dynamics simulations of shock waves (see, for example, Ref. [5]) propagating through crystalline NaCl. Such commonly utilized simulations solve the classical equations of motion for atoms subject to an empirically-constructed interaction potential and incorporate thermal effects and deformation of the crystal lattice. In these calculations, planar shock waves are generated within 3D computational cells of perfectly crystalline atoms with cross section 17×17nm and length in the shock propagation direction ranging from 158nm to 235 nm (2-3 million atoms) at T=4.2K. Due to periodic boundary conditions in the directions transverse to the propagation direction, an infinitely planar shock propagates away from the constrained atoms into the the cell which is oriented along either the [111] or [100] directions of NaCl. We have performed simulations with computational cell cross-sections of up to 135×135 nm and obtained results in agreement with cross sections of 17×17 nm which indicates that the computational cell sizes are large enough that periodic boundary conditions do not play an artificial role in the behavior of the shocked material. Atomic interactions are treated using unit charge Coulomb interactions combined with Lennard-Jones interactions. These potentials are found to yield a lattice

constant (5.64 Å) in agreement with the experimental value and to yield sound speeds that deviate from experimental measurements by 10-20%.

These molecular dynamics simulations do not explicitly solve Maxwell's equations for the electric and magnetic fields. However, since the wavelength of radiation emitted at frequencies considered here (longer than 10 μm) is much longer than the dimensions of the computational cell, it is expected that the total polarization current generated in the computational cell will be closely related to the generated electromagnetic radiation for frequencies above the phonon frequencies (above about 10 THz in this NaCl model) where the material has good transmission properties. The shock propagation direction component of the total electric polarization current is $J = \sum_i v_{z,i} q_i$ where q_i is the charge and $v_{z,i}$ is the shock direction (z) component of the velocity of atom *i*. Emission characteristics are discussed in detail below.

Figure 1 shows results of about 30 ps duration simulations of shocks propagating in the [111] and [100] directions initiated with piston velocities of 200 m/s. This relatively small piston velocity generates a shock that applies a uniaxial strain of 0.03-0.04 to the post-shock material and increases the material temperature less than 1K. Figure 1 compares the shocked and unshocked Fourier transform of the shock propagation direction component of the total electric polarization current in the computational cell. Narrow peaks are observed in the shocked simulation that do not exist in the unshocked simulation. From the peak widths, the coherence length of the radiation emitted in vacuum is determined to be about 5mm and 3mm for the 16 THz, [111] peak and 22 THz, [100] peak, respectively. These lengths are comparable to those of some commonly used lasers. The coherence times are nearly Fourier transform-limited, suggesting that longer coherence lengths could be demonstrated by increasing the shock propagation time.

Equation 3 predicts that emission should occur in multiples of 5.4 THz in the [111] case since the periodic unit for the [111] direction in NaCl $a = 9.78$ and the shock speed observed in the simulation is $v_s = 5300 m/s$. The 16 THz and 32 THz peaks on the left of Figure 1 correspond to 3 and 6 times the fundamental frequency of 5.4 THz ($\ell = 3$ and $\ell = 6$ in Equation 3), in excellent agreement with theory (gray arrows). The 16 THz peak can be attributed

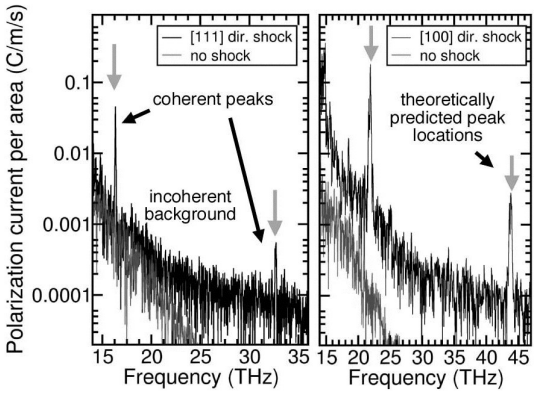

FIGURE 1. Fourier transform of the electric polarization surface current component in the shock propagation direction for molecular dynamics simulations of a shock propagating through NaCl in the [111] direction (left) and [100] direction (right). Narrow bandwidth, coherent peaks exist in the shocked simulations (black) that do not exist in the simulations without shocks. Gray arrows are emission frequencies predicted by Eq. 3. The coherence length for emission from the 16 THz peak in the [111] case is about 5 mm, comparable to that of some commonly used lasers. Thermal noise gives rise to an incoherent background.

to structure within the unit cell of distance $a = 3.26$ (i.e. $\ell = 1$ if $a = 3.26$ in Eq. 3) which is the distance between atomic lattice planes of like charge in the [111] direction (the NaCl crystal consists of alternating planes of positively and negatively charged atoms in the [111] direction.) Lattice planes are compressed as the shock propagates through the crystal, generating an alternating polarization current with a frequency associated with the rate at which the shock propagates through the lattice planes. If the shock speed is constant, the generated frequencies are constant and the coherence time of the emitted radiation is expected to be proportional to the time duration of the propagation of the shock wave. Equation 3 is also in good agreement with the [100] direction data where $a = 5.64$ and the observed shock speed is $v_s = 6200 m/s$ with peaks corresponding to the $\ell = 2$ and $\ell = 4$ cases. These peaks correspond to the distance between neighboring lattice planes in the crystal.

The power and spatial distribution of the emitted radiation can be simply estimated in the specific case where the shock front diameter is much less than the wavelength of the coherently emitted radiation. In

this case, the propagating shock wave acts as a point source of radiation emitting a dipole-like distribution of radiation (peaked in the plane of the shock wave) with power,

$$p(\omega_0) = \frac{1}{4\pi\varepsilon_0}\frac{j_0^2 A^2 \omega_0^2}{3c^3} \qquad (4)$$

where j_0 is the polarization current per unit area on the shock front surface (plotted in Figure 1) and A is the shock front area. For a $5\mu m$ diameter shock front (e.g., a laser-driven shock) and $j_0 = 0.18$C/s/m for the 22 THz peak on the right side of Figure 1, Equation 4 predicts the power coherently radiated at 22 THz is 3×10^{-11} Watts while the shock propagates. Collecting and focusing this radiation can yield electric field amplitudes up to about 0.1 V/cm. We find that the power radiated can be increased by orders of magnitude when larger shock front areas and deviations from perfect planarity considered. [6]

The emission peaks studied in this work have frequencies that are up to two times higher than phonon frequencies. Observation of these peaks requires that there be spectral components of atom velocities at these higher frequencies around the shock front. Such high frequency components of atomic motion can be generated only if the shock front (or part of it) is very sharp. The natural shock front rise distances in the molecular dynamics simulations are several lattice planes. This effect represents a mechanism through which elastic shock wave rise times can potentially be experimentally measured; to our knowledge, such a measurement has never been made.

CONCLUDING REMARKS

We have shown that a mechanical shock wave propagating through a crystalline polarizable material can produce coherent radiation. The key to this process is the periodicity of the crystal lattice. Suspected non-equilibrium (non-thermal) optical radiation in the visible regime has been observed from shocked dielectrics under some conditions, [7, 8, 9, 10] but this radiation appears to have a different origin than the coherent emission discussed here which occurs in a different part of the electromagnetic spectrum and results from the crystallinity of the lattice.

We note that it is well-known that coherent radiation at almost any frequency can be obtained from a *coherent* source using materials with a nonlinear optical response. This approach utilizes radiation from a coherent source to generate new coherent radiation. The coherent radiation mechanism presented in this work is fundamentally distinct from such nonlinear approaches in that coherence results from the crystal lattice rather than another coherent source. The optical nonlinearity mechanism works in amorphous materials while the shocked crystal mechanism does not.

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